Area-dependence of spin-triplet supercurrent in ferromagnetic Josephson junctions

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Area-dependence of spin-triplet supercurrent in ferromagnetic Josephson junctions

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Josephson junctions containing multiple ferromagnetic layers can carry spin-triplet supercurrent under certain conditions. Large-area junctions containing multiple domains are expected to have a random distribution of 0 or π coupling across the junction surface, whereas magnetized samples should have uniquely π coupling everywhere. We have measured the area dependence of the critical current in such junctions, and confirm that the critical current scales linearly with area in magnetized junctions. For as-grown (multi-domain) samples, the results are mixed. Samples grown on a thick Nb base exhibit critical currents that scale sub-linearly with area, while samples grown on a smoother Nb/Al multilayer base exhibit critical currents that scale linearly with area. The latter results are consistent with a theoretical picture due to Zyuzin and Spivak that predicts that the as-grown samples should have global $\pi/2$ coupling.

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I. INTRODUCTION

When a superconducting (S) metal is placed in contact with a nonsuperconducting (N=normal) metal, Cooper pairs can "leak" out of the superconductor and modify the properties of both materials. This process, called the superconducting proximity effect, has been studied for several decades.[1] When the normal metal is replaced by a ferromagnetic (F) metal, the two electrons of the pair enter different spin bands with different Fermi wavevectors. As a result, the pair correlations oscillate and decay rapidly with increasing distance from the S/F interface.[2] The oscillating, short-ranged proximity effect in S/F systems was predicted as early as 1982,[3] and has been observed convincing by many groups over the past decade.[4]

In 2001, it was predicted that pair correlations with spin-triplet symmetry can be generated in S/F systems containing certain kinds of magnetic inhomogeneities involving non-collinear magnetizations, even if all of the superconducting materials in the system have conventional spin-singlet symmetry.[5–8] Spin-triplet pair correlations do not experience the exchange field in F, hence the proximity effect due to those pair correlations is long ranged. Some experimental evidence for spin-triplet correlations appeared in 2006,[9,10] then more convincing evidence appeared in 2010.[11–14] Our own contribution[11] was based on measurements of the critical current I_c in Josephson junctions of the form S/F'/SAF/F"/S, where SAF stands for "synthetic antiferromagnet" and F' and F" are thin ferromagnetic layers whose magnetizations must be at least partly non-collinear with that of the SAF. The SAF is a Co/Ru/Co trilayer with Ru thickness 0.6 nm, which causes anti-parallel coupling of the two surrounding Co magnetizations. Because of that anti-parallel coupling, the SAF produces nearly zero net magnetic flux, which would otherwise distort the "Fraunhofer patterns" one observes in plots of I_c vs magnetic field H applied in the plane of the Josephson junction. We found that I_c in our samples hardly decreased as the total Co thickness was increased up to 30 nm, whereas I_c decayed rapidly in similar samples without the F' and F" layers.[15] The long-range nature of the supercurrent in the samples containing F' and F" layers provided strong evidence for the spin-triplet symmetry of the current-carrying electron pairs. Furthermore, we have recently shown that I_c increases further when we magnetize our samples with an in-plane applied magnetic field.[16] The explanation is that the F' and F" layers are magnetized parallel to the field, while the SAF undergoes a "spin-flop" transition whereby the two Co layers end up with their magnetization perpendicular to the direction of the applied field.[17,18] According to theory,[19–22] this configuration with perpendicular magnetizations maximizes the magnitude of the spin-triplet supercurrent.

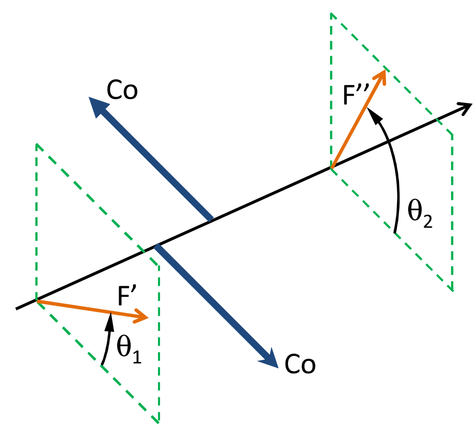

FIG. 1: Cartoon showing relative orientations of magnetization for the ferromagnetic layers in our Josephson junctions. If angles θ_1 and θ_2 have the same sign (where we constrain $|\theta_1|, |\theta_2| < \pi$), the junction will have π coupling; if they have opposite signs, the junction will have 0 coupling.

The results described above raise several questions, only some of which have been answered by subsequent work in our group.[16,23] The question that motivated this paper arises from the theoretical prediction that Josephson junctions of the form S/F'/F/F"/S, carrying spin-triplet supercurrent, can be either in the 0-state or the π-state depending on the relative orientations of the three ferromagnetic layers.[19,20] (It does not matter whether the central F layer is a single ferromagnetic layer or an SAF.[21,22]) The situation is illustrated in Figure 1, which shows the relative orientations of the magnetizations of all four ferromagnetic layers in our junctions. According to theory, if the two angles θ_1 and θ_2 have the same sign, then the junction will have π coupling; if they have opposite signs, the junction will have 0 coupling.[20–22] (We define the angles by the constraint $|\theta_1|, |\theta_2| < \pi$.) Since the magnetic layers in our samples consist of many domains when the samples are first grown, we would expect the Josephson coupling in our junctions to exhibit a random spatially-varying pattern of 0-coupling and π-coupling across the junction area. In that case, if a fixed difference in gauge-invariant phase is applied across the junction, some areas of the junction will provide positive supercurrent while others will provide negative supercurrent – i.e. supercurrent flowing in the opposite direction. One could then calculate the total supercurrent naively using an analogy to the random walk problem: while the mean supercurrent averaged over many domains would be zero, the typical supercurrent in a given sample would be proportional to the square-root of the number of domains, hence to the square-root of the junction area. After the samples are magnetized, the magnetizations of the F' and F" layers are parallel to each other, hence θ_1 and θ_2 have the same sign and the junction should have π-coupling everywhere. In that case, the critical supercurrent will be proportional to the junction area, as is the case in conventional Josephson junctions.

A completely different view of a Josephson junction containing a random spatially-varying pattern of 0 and π couplings has been proposed by Zyuzin and Spivak (ZS).[24] Those authors addressed S/F/S junctions with spin-singlet rather than spin-triplet supercurrent, and considered the situation where the F-layer thickness is large, so that the average supercurrent is small, whereas mesoscopic fluctuations of the Josephson coupling have random sign. (Spatially-varying 0 and π couplings for spin-singlet supercurrent could also be produced by fluctuations in F-layer thickness, as discussed in Ref. [25].) In our samples, the spin-singlet supercurrent is negligibly small (see Figure 3 in Ref. [11]), and the random-sign spin-triplet Josephson coupling arises from the local variations in magnetic domain structure. In spite of the different mechanisms underlying the spatially-varying random-sign Josephson coupling, there is no apparent reason why the ZS model should not apply to our spin-triplet Josephson junctions. ZS calculated the total energy of such a junction, and concluded that the ground state corresponds to, on average, a $\pi/2$ phase difference between the two superconducting electrodes. The phase difference is spatially modulated, with local variations toward lower phase difference in regions of 0-coupling and larger phase difference in regions of π-coupling. According to the ZS result, the total supercurrent scales with the junction area, as is the case for conventional Josephson junctions.

The purpose of this paper is to measure experimentally the area dependence of the supercurrent in our Josephson junctions, to determine which of the pictures presented above applies.

II. SAMPLE FABRICATION

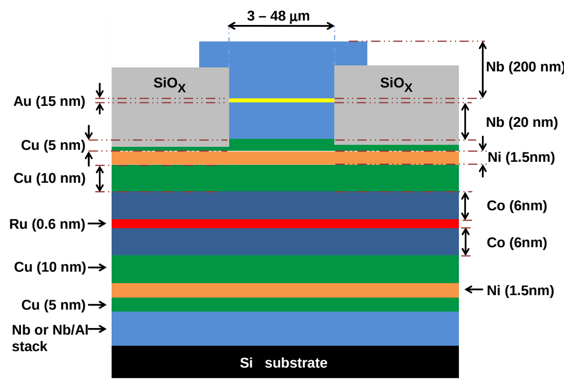

FIG. 2: (color online) Schematic diagram of Josephson junction samples (not to scale). The current flows in the vertical direction.

The Josephson junctions in this work were fabricated by dc triode sputtering, photolithography and ion-milling, as described in our previous publications.[11,15] The structure of the junctions is shown schematically in Figure 2. In this work, we have grown two types of samples with different superconducting base layers: either a single 150-nm layer of Nb, or a Nb/Al multilayer stack described below. For the first kind of sample we start by growing a multilayer of the form Nb(150)/Cu(5)/Ni(1.5)/Cu(10/Co(6)/Ru(0.6)/Co(6)/Cu(10)/Ni(1.5)/Cu(5)/Nb(20)/Au(15), where all thicknesses are in nm. That stack is sputtered in one run without breaking vacuum. For the second type of sample, the Nb base layer was replaced with a Nb/Al multilayer of the form [Nb(40nm)/Al(2.4nm)]$_3$/Nb(40nm)/Au(15nm). This structure was motivated by the work of Thomas et al.[26], who used a thin Nb/Al multilayer on top of thick Nb to reduce surface roughness. For those samples, the chamber was opened briefly after the Au deposition, while flowing N_2 gas, to change sputtering targets. (Our sputtering system holds 6 targets, whereas the second type of samples require 7 different materials.) After pumpdown, we sputtered first 20 nm of Nb, followed by the rest of the stack up through the top Au layer. We know from our top electrode fabrication procedure that the top 15-nm Au layer adequately protects the underlying Nb from oxidation, and is driven superconducting when sandwiched between two Nb layers. The same should be true for the bottom Au layer protecting the

Nb/Al multilayer base.

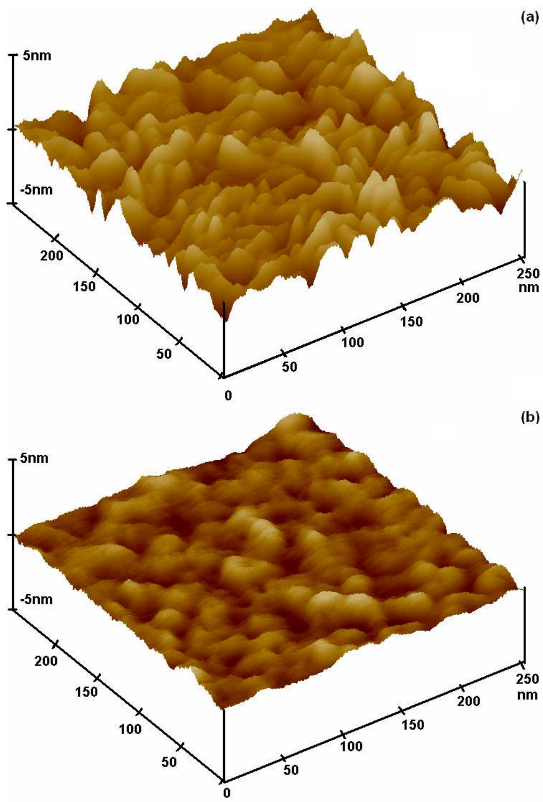

FIG. 3: (color online) Atomic force microscopy pictures of (a) a 200-nm thick Nb base layer and (b) a Nb/Al multilayer as described in the text.

To ascertain the surface roughness of the two types of base layers, we performed atomic force microscopy (AFM) measurements on a bare 200-nm Nb base and on a Nb/Al multilayer stack (up to the Au layer discussed above). The results are shown in Figure 3. The root-mean-squared roughnesses of the first and second base layers are 0.53 nm and 0.23 nm, respectively, over the $250 \times 250\text{nm}^2$ area shown. As expected, the Nb/Al multilayer provides a smoother base than the pure Nb.

For both types of samples, Josephson junctions with circular cross section were defined by photolithography and ion milling. Because we wanted to obtain data on multi-domain samples covering a large dynamic range of areas, we fabricated junctions with diameters of 3, 6, 12, 24, and 48 micrometers. Unfortunately, the largest samples rarely produced high-quality data, hence we restrict ourselves here to the samples of diameters 3, 6, and 12 μm. Another difference between the samples measured here and those measured in our previous publications is that these were ion milled only to the top copper layer in order to keep the magnetic domain structure intact, whereas our previous samples were typically ion milled partway through the top Co layer.

III. EXPERIMENTAL RESULTS

All of the data reported here were acquired at 4.2K with the sample dipped into a liquid helium dewar. A current comparitor circuit using a Superconducting Quantum Interference Device (SQUID) as a null detector was used to measure the current-voltage (I-V) characteristic of the samples.[27] All samples exhibit the standard I-V characteristic for an overdamped Josephson junction:

$$V(I) = Sign[I] * R_N \mathrm{Re}[(I^2 - I_c^2)^{1/2}] \quad (1)$$

where R_N is the normal-state resistance determined from the slope of the V-I relation at large currents.

A. Fraunhofer patterns and result of magnetizing samples

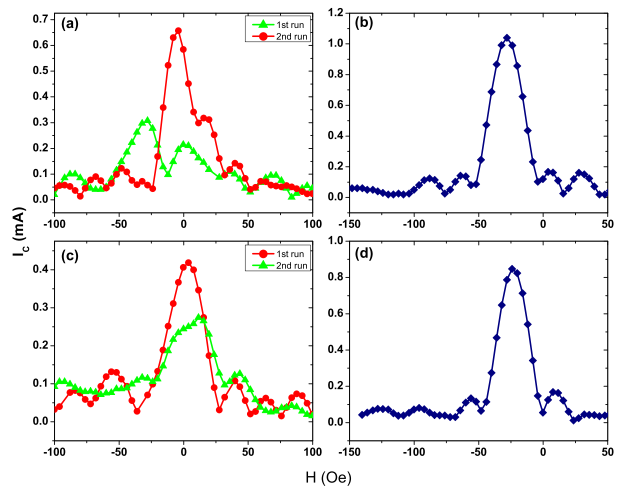

FIG. 4: (color online) Critical current vs. applied magnetic field for two 3-μm diameter Josephson junctions, measured in the virgin state (panels a and c), and after the samples were magnetized by an in-plane field of 2 kOe (panels b and d). In the virgin state, two separate runs are shown for each sample. The lines connect the data points; they are only guides for the eye.

All samples are initially characterized by applying a small magnetic field perpendicular to the current direction, i.e. in the plane of the substrate. A plot of I_c vs H should yield the classic "Fraunhofer pattern" (actually an Airy pattern for our circular pillars). Figure 4 shows representative I_c vs H data for two 3-μm-diameter junctions, in the virgin state (panels a and c), and after being magnetized by an in-plane field of 2 kOe (panels b and d). We measured a few samples several times to determine how much the data vary from run to run; panels a and c show two virgin-state runs for two of these samples. Several features are evident from the data: i) the Frauhofer patterns in the virgin state fluctuate from run to run; ii) the quality of the Fraunhofer patterns is better after the samples are magnetized than in the virgin state; iii) I_c is enhanced after the samples are magnetized, as we reported recently;[16] and iv) the central peak in the Fraunhofer patterns of the magnetized samples is shifted to negative field by about 30 Oe.

The variability and relatively low quality of the Frauhofer patterns in the virgin state are undoubtedly due to the random domain structures of the ferromagnetic layers in the samples.[28,29] One cannot know *a priori* if the problem is due to the 1.5-nm thick Ni F' and F" layers or to the two 6-nm thick Co layers making up the SAF. We believe it is the former, given the improvement in the quality and reproducibility of the Fraunhofer patterns after magnetization. Magnetizing the samples forces the Ni domain magnetizations to point in nearly the same direction, and probably causes the average domain size to grow. In contrast, the Co/Ru/Co SAF is expected to have its best antiparallel coupling in the virgin state.

The enhancement of I_c by magnetizing the samples has potential contributions from two factors. In our recent work,[16] we emphasized optimization of the angles θ_1 and θ_2 between the inner and outer ferromagnetic layer magnetizations. Those angles vary randomly across the junction area in the virgin-state samples, whereas they should both be close to the optimal value of $\pi/2$ after the samples are magnetized, due to the Co/Ru/Co SAF undergoing a spin-flop transition. A second contributor may be the fact that, in the virgin state, the Josephson coupling varies randomly between 0-coupling and π-coupling across the junction area. If the random-walk picture discussed earlier is valid, then one would expect the value of I_c in a typical sample to scale with the square-root of the junction area, as discussed earlier. After the samples are magnetized, there should be π-coupling everywhere, so that the supercurrent adds constructively across the entire junction area.

Finally, the shift in the Fraunhofer patterns after magnetization has been observed previously.[29–31] The central peak occurs at the point where the flux due to the applied field exactly cancels the intrinsic flux due to the magnetization of the Ni layers inside the junction.

The evolution of I_c as the sample is magnetized is shown for a 6-μm diameter Josephson junction in Figure 5. The sample was first measured in the as-grown state (H = 0). Then the magnetizing field H was stepped up to 3600 Oe with varying step sizes evident in the figure. After application of each value of H, the field is reduced to zero and the Fraunhofer pattern is measured in low field. The squares show the resulting values of I_cR_N as the sample is magnetized. For low fields, nothing happens. Then there is a shallow dip in I_cR_N for H near 500 Oe. That dip is observed in many samples, but is not fully understood; it may be related to a change of the Ni domain structure. When H is increased above 500 Oe, I_c increases sharply. The field range where I_c increases corresponds to the field range where the Ni films become magnetized. (We know this because the field where I_c increases varies with Ni layer thickness, and matches the coercive field of the Ni determined from separate magnetization measurements of large-area Ni films.[16]) Figure 5 also shows what happens when a field is applied in the opposite direction to the original magnetizing field (circles). Again, nothing happens for small field values. Then, as the Ni films are demagnetized, I_c drops to values as low as or even lower than the value at the dip we observed when first magnetizing the samples. As the Ni films are re-magnetized in the negative direction, I_c increases sharply again to a value essentially identical with that observed on the positive field side. We have measured full magnetization curves for several samples, and they all look very similar to the one shown in Figure 5.

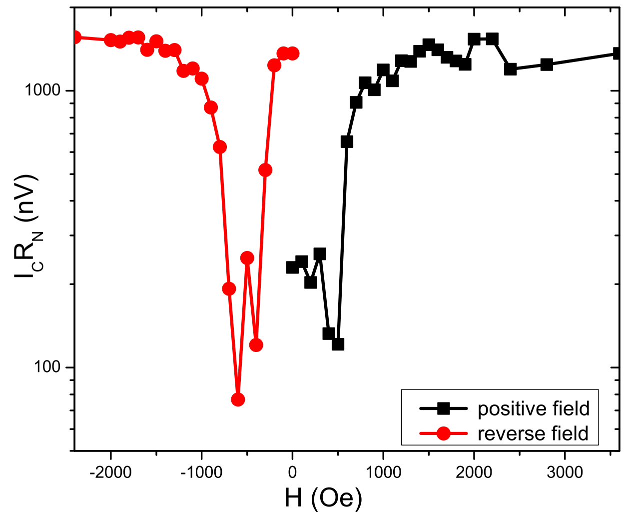

FIG. 5: (color online) I_cR_N product vs. applied in-plane field for a 6-μm diameter Josephson junction. The sample was first magnetized in positive field (squares), then the sample was demagnetized and finally re-magnetized in negative field (circles).

B. Area dependence: samples with Nb base layer

To provide good statistics and to reveal the extent of sample-to-sample fluctuations, we have fabricated and measured a large number of Josephson junctions with diameters of 3, 6, and 12 μm. Figure 6 shows the results for samples grown on our traditional thick (150 nm) superconducting Nb base, for both the virgin and magnetized states. The points representing the magnetized state (open symbols) are averages of the measurements taken after application of 1600, 2000, and 2400 Oe. (There is little variation of I_cR_N between those three measurements.) The points representing the virgin state (solid symbols) are usually averaged over two runs, although a few samples were measured only once, and one sample was measured 5 times in the virgin state.

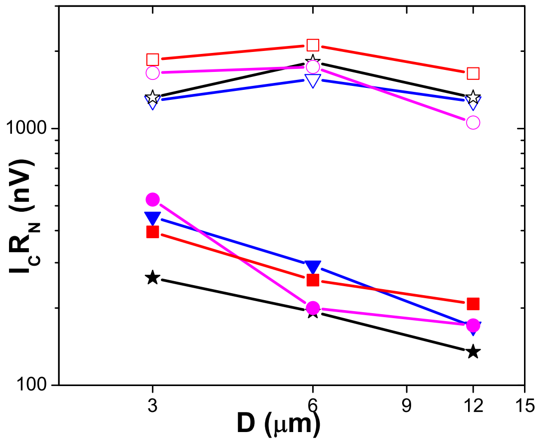

FIG. 6: (color online) Critical current times normal-state resistance vs. junction diameter for Josephson junctions grown on a 150-nm Nb base electrode. Solid symbols represent virgin-state data; open symbols represent data acquired after the samples were magnetized by a large (≈2000 Oe) in-plane magnetic field.

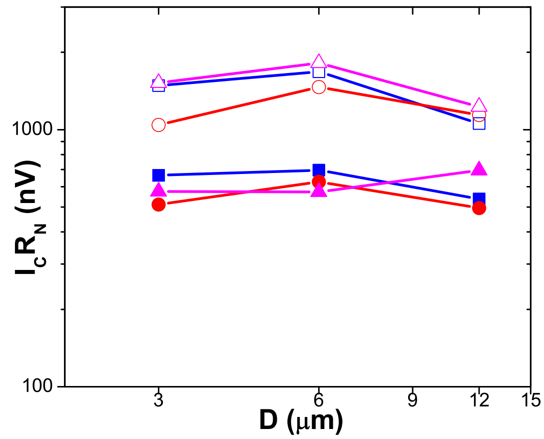

FIG. 7: (color online) Critical current times normal-state resistance vs. junction diameter for Josephson junctions grown on a Nb/Al multilayer as described in the text. Solid symbols represent virgin-state data; open symbols represent data acquired after the samples were magnetized by a large (2000 Oe) in-plane magnetic field.

The results of the magnetized state measurements in Figure 6 clearly show that I_cR_N is essentially independent of sample area. Since R_N is inversely proportional to junction area, this means that I_c is proportional to area. That is the usual situation, and is what one expects when the Josephson coupling is uniform across the junction area. In contrast, the virgin-state data show a decrease in I_cR_N with increasing sample size. According to the random walk model discussed in the introduction, I_c should scale with the square-root of the junction area, hence I_cR_N should scale inversely with the square-root of area, or equivalently, inversely with junction diameter D. The virgin-state data shown in Figure 6 do exhibit a noticeable decrease with junction diameter, supporting the random walk picture, although the dependence is slightly less steep than $I_cR_N \propto D^{-1}$.

C. Samples with Nb/Al base layer

As shown in Figure 3, the Nb/Al multilayer base provides a smoother surface than the pure Nb base layer. We were curious as to whether the smoother base would influence any of the Josephson junctions properties. We performed the same measurements of I_c on the second batch of samples, grown on the smoother Nb/Al base, as were performed on the first batch, grown on pure Nb. The results are shown in Figure 7. In the magnetized state, the results agree closely with those of the first batch, shown in Figure 6. Not only is I_cR_N essentially independent of junction diameter, but the actual values of I_cR_N are very close to those of the previous batch of samples. After being magnetized, there is remarkable consistently in the values of I_cR_N for the 21 samples displayed in Figures 6 and 7. In the virgin state, however, the story is different. In contrast to what we observed in the first batch of samples, I_cR_N in the second batch hardly varies with junction diameter. These samples deposited on top of the smoother base electrode appear to validate the ZS theory, which predicts that I_c should scale linearly with junction area.

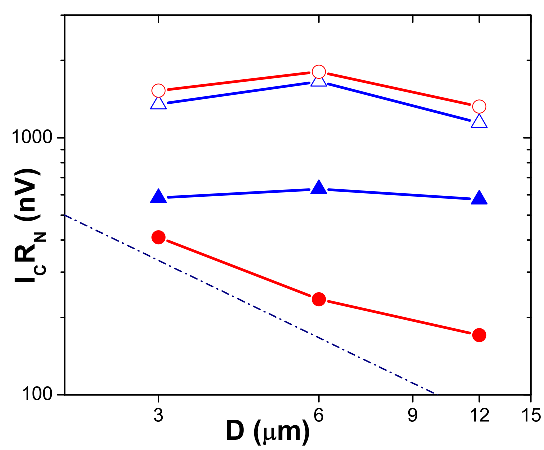

FIG. 8: (color online) Summary of I_cR_N data for all the Josephson junctions studied in this work. Each symbol represents the average value for all samples of a given size and base layer, in either the virgin-state (solid symbols) or after being magnetized (open symbols). The circles represent samples grown on a 150-nm Nb base layer, while the triangles represent samples grown on a Nb/Al multilayer described in the text. The dot-dashed line illustrates the relation $I_cR_N \propto D^{-1}$.

The situation is summarized in Figure 8, where we have averaged together the values of I_cR_N for all samples of a given diameter, fabricated on a given base layer. With some of the sample-to-sample fluctuations averaged out, the trends are striking: i) The magnetized data are remarkably consistent, and hardly depend on the base layer; ii) the virgin-state data from samples deposited on the thick Nb base exhibit I_cR_N values that decrease substantially with junction diameter, but not quite as fast as D^{-1}, which is shown by the dot-dashed line in the figure; iii) the virgin-state data from samples deposited on the smoother Nb/Al multilayer base exhibit I_cR_N values that are independent of junction diameter.

IV. DISCUSSION

The results summarized in Figure 8 imply that the roughness of the base layer has a profound effect on the area scaling of I_c in the virgin state. One can imagine several possible explanations for this observation.

One possible explanation is that the spectrum of Andreev bound states, and hence the Josephson current, in an S/F/S junction depends in a fundamental way on whether the S/F interface is smooth or rough. There has been some theoretical work on interface roughness in S/N systems,[32] but we are not aware of any such work that addresses our results directly.

A second possibility is that the roughness of the Nb base layer perturbs the domain structure of the Ni F' and F" layers – possibly even to the extent that one or both of those layers are not continuous. It is well known that many magnetic materials exhibit magnetically "dead" layers when placed next to nonmagnetic or superconducting materials.[33–35] Dead layers can arise even at a perfect interface due to electronic structure effects;[36] they are typically exacerbated by interface roughness or by interdiffusion between the two metals near the interface. In cases when surface roughness is dominant, one can imagine the presence of isolated magnetic or superparamagnetic clusters that are only weakly coupled to the bulk of the magnetic film; in the latter case one would expect large spin fluctuations to be highly detrimental to superconductivity.

Regarding the potential role of magnetically dead layers in our experiments, we have estimated the magnetic dead layer thickness for our sputtered samples at the Ni/Cu interface to be 0.25 nm per interface at a temperature of 5 K (although the multilayer samples used for that study were grown directly on the Si substrate rather than on a thick Nb base).[37] We also know that the coercive fields of the Ni layers in our Josephson junctions increase continuously with decreasing Ni thickness down to 1 nm, as shown in the data in Figure 2 of Ref. [16], so the rough base does not induce any globally anomalous magnetic behavior for Ni films at least 1 nm thick. In addition, there is evidence for generation of some spin-triplet supercurrent even in samples containing F' and F" Ni layers only 0.5 nm thick,[23] indicating the presence of at least some ferromagnetic regions in those very thin layers. More importantly, one still has to explain how rough or discontinuous F' and F" layers would affect the area scaling of the junction critical current in the virgin state. If the magnetically dead regions take the form of a discontinuous network of superparamagnetic or nonmagnetic clusters, then the Josephson junctions should be modelled as a random network of 0-coupled, π-coupled, and uncoupled regions. As long as the correlation length of those regions is much less than than the Josephson penetration length, λ_J, then the Zyuzin-Spivak model should apply. In our junctions, we estimate λ_J to be of order 10 μm in the virgin state, while the typical domain size in the Co layers is of order 1-3 μm,[16] and perhaps smaller in the Ni layers, and the correlation length of the roughness is estimated from Figure 3 to be 20-30 nm.

Finally, a third possible explanation (and the least interesting) is that the observed sub-linear scaling of the supercurrent with junction area for the junctions grown on the rougher Nb base is simply a reflection of the gradual deterioration of the quality of the Fraunhofer patterns with increasing sample size. A possible way to ameliorate that issue in the future would be to use PdNi alloy rather than pure Ni as the F' and F" layers. PdNi is a weak ferromagnetic material with small magnetization, and in earlier work we were able to produce Josephson junctions with high-quality Fraunhofer patterns even with much thicker PdNi layers than one would need for this experiment.[29] The optimal PdNi thickness for producing spin-triplet supercurrent is in the range of 4-6 nm,[11] which is much thicker than the 1-2 nm optimal range for Ni,[23] but still much less than the PdNi layers in our earlier work.[29] We chose pure Ni for the F' and F" layers in the present work because it produces the largest values of I_c in the virgin state,[23] but thicker PdNi layers might be less sensitive to the nm-scale roughness of the Nb base.

V. CONCLUSIONS AND OUTLOOK

In summary, we have measured the area-dependence of the critical current in S/F/S Josephson junctions carrying spin-triplet supercurrent. After the samples are magnetized, the critical current has its largest value, and it scales linearly with area as is the case for conventional Josephson junctions. In the virgin state, however, the results are mixed. Samples grown on our traditional thick Nb base exhibit critical currents that grow sub-linearly with area, whereas samples grown on a smoother Nb/Al multilayer base exhibit critical currents with conventional area scaling. The former may be an indication that the supercurrent is not uniform over the junction area, while the latter provides indirect support for a theoretical model of Zyuzin and Spivak.[24]

In the future, it would be interesting to measure the current-phase relation of our junctions, both in the virgin and magnetized states. According to the ZS theory,[24] the virgin state junctions should be in a $\pi/2$ state, while according to spin-triplet junction theory,[20–22] the magnetized junctions should be in the π state. Current-phase measurements are technically more challenging than the critical current measurements reported here, but they might provide more direct evidence of the underlying physics than do the area-dependent measurements reported area.

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